

UNIVERSITÀ DEGLI STUDI DI PADOVA

Dipartimento di Fisica e Astronomia
"Galileo Galilei"



Corso di Laurea in Fisica

TESI DI LAUREA

Studio ed analisi dei livelli di radioattività in un ciclotrone da 70 MeV

Relatore: Dott.ssa Laura De Nardo

Correlatore: Dott.ssa Lucia Sarchiapone

Laureanda: Bianca Giacomelli

Anno Accademico 2015/2016

Sommario

Lo scopo di questo lavoro di tesi è lo studio della radioattività indotta dall'interazione di un fascio di protoni, accelerato dal ciclotrone 70p ai Laboratori Nazionali di Legnaro, nei costituenti e zone limitrofe dell'acceleratore stesso. Sia misure dirette che metodi analitici di previsione sono stati usati per quantificare la radioattività indotta dal fascio di protoni a 70 MeV, durante i test di commissioning del ciclotrone stesso, in tre campioni: 1) un bersaglio spesso di rame utilizzato per i test di estrazione del fascio di protoni a $1\mu A$ all'interno del vano del ciclotrone, 2) un campione dell'acqua di raffreddamento di una sonda di rame usata per test di iniezione del fascio a $1\mu A$ e 3) un campione di polvere proveniente dalla cave dell'acceleratore e attivato in corrispondenza della condotta della linea del fascio durante l'irradiazione, con il fascio di protoni a 70 MeV e $100\mu A$, di un bersaglio di rame schermato con polietilene e piombo posto nel bunker di irraggiamento. Il fine di questa tesi è sia di quantificare in maniera operativa la radioattività indotta nei campioni considerati e quindi derivarne la dose equivalente, che di applicare alcuni metodi analitici per ottenere una stima orientativa di entrambe le grandezze.

I metodi analitici usati sono stati infatti intesi come potenziali strumenti utili alla stima dell'ordine di grandezza della radioattività indotta e quindi della dose equivalente, in assenza, ma non in sostituzione, di stime prodotte da simulazioni al computer.

Abstract

The purpose of this thesis work is to study nuclear activation processes due to the interaction of a proton beam in the materials of the 70p cyclotron accelerator at LNL¹, its irradiation cave and beam line pipe. Experimental measures and analytical methods were used to quantify the radioactivity induced, during the accelerator commissioning tests, in three samples: 1) a thick copper target, or beam dump (BD), used in low current tests for beam extraction at 70 MeV and $1\mu A$, 2) a sample of cooling (DDT) water of a copper beam interceptor used for beam injection tests at $1\mu A$ and 70 MeV, and 3) a sample of the dust from the accelerator irradiation cave placed close to the beam line pipe during the proton irradiation at 70 MeV and $100\mu A$ of a copper target shielded with lead and polyethylene. The results of this work aim both to operatively quantify the proton beam induced activity in the mentioned samples and to apply some analytical methods for a rough appreciation of the magnitude of the radiation and respective equivalent dose.

The analytical methods studied are not intended to replace computer simulations of energetic hadronic cascades, however they can constitute a useful tool to make a first guess of the magnitude of the problem also in the eventual absence of estimates provided by computer simulations.

¹National Legnaro Laboratories

Contents

| | | |
|----------|---|-----------|
| 1 | Cyclotron radiation detection | 9 |
| 1.1 | The SPES facility and the 70 MeV Cyclotron | 9 |
| 1.1.1 | Radiation fields associated with particle accelerators | 10 |
| 1.2 | Gamma Detectors | 11 |
| 2 | Prediction of induced activity in target and dumps | 13 |
| 2.0.1 | Hadron Cascades | 13 |
| 2.1 | Induced activity in a copper beam dump by hadron nuclear interactions . | 15 |
| 2.1.1 | Sullivan Overton approximation | 16 |
| 2.1.2 | Radioactivity in copper beam dump from the 70 MeV beam proton nuclear interactions | 18 |
| 2.1.3 | Radioactivity in copper targets from secondary neutron nuclear reactions | 19 |
| 2.1.4 | Radioactivity in copper targets from neutron <i>radiation capture</i> . . | 20 |
| 2.2 | Radioactivity induced in the cyclotron cooling water | 21 |
| 2.3 | Induced activity in dust near the beam line pipe in the irradiation cave . . | 24 |
| 3 | Measurement campaign and collected data | 27 |
| 3.1 | Conclusions | 29 |
| | Bibliography | 30 |

Introduction

The operation of particle accelerators causes induced activity in its components and surroundings that can constitute a permanent hazard to staff and also limit its performance: a realistic appreciation of the magnitude of the radiation levels is therefore an essential requirement for a safe and efficient accelerator operation. Energetic hadron interactions with the accelerator components, and also subsequent production of secondary radiation, determine an induced activity in the materials that persists after the irradiation time, or switching off of the accelerator.

The purpose of this thesis work is to study nuclear activation processes due to the interaction of a proton beam in the materials of the 70p cyclotron accelerator at LNL², its irradiation cave and beam line pipe. Experimental measures and analytical methods were used to quantify the radioactivity induced, during the accelerator commissioning tests, in three samples: 1) a thick copper target, or beam dump (BD), used in low current tests for beam extraction at 70 MeV and $1\mu A$, 2) a sample of cooling (DDT) water of a copper beam interceptor used for the proton beam injection tests at $1\mu A$ and 70 MeV, and 3) a sample of the dust from the accelerator irradiation cave placed close to the beam line pipe during the proton irradiation at 70 MeV and $100\mu A$ of a copper target shielded with lead and polyethylene. The results of this work aim both to operatively quantify the proton beam induced activity in the mentioned samples and to apply some of the analytical methods for a rough appreciation of the magnitude of the radiation and respective equivalent dose.

The analytical methods studied are not intended to replace computer simulations of energetic hadronic cascades, however they can constitute a useful tool to make a first guess of the magnitude of the problem also in the eventual absence of estimates provided by computer simulations.

This thesis is structured in three chapters.

- The first chapter consists of an introduction to the SPES facility, the 70 MeV cyclotron, the gamma detector used to measure the activities and to the contextualization of the samples study in the commissioning of the 70p cyclotron at the LNL.
- In the second chapter, an overview of the main nuclear interaction processes between high and medium energetic hadrons and target nuclei is followed by the

²National Legnaro Laboratories

analytical estimates of the induced activities and derivation of the equivalent dose in the copper beam dump and the cooling water sample. A qualitative outline of the expected radioisotopes in the dust (from the cyclotron irradiation cave) placed close to the beam line pipe concludes the chapter.

- In the third chapter are presented the experimental data collected during the measurement campaign. It follows a brief comparison between the dose rates computed from the measured activities and the dose rates estimated through analytical methods. The chapter closes with an evaluation of the dose rates obtained by experimental measures of the samples activities.

Chapter 1

Cyclotron radiation detection

1.1 The SPES facility and the 70 MeV Cyclotron

The SPES (Study for the Production of Exotic Species) project aims to get high intensity and quality beams of neutron-rich nuclei to implement research in nuclear structure, reaction dynamics and interdisciplinary fields like medical, biological and material sciences. The driver of the SPES project is a cyclotron, able to provide two simultaneous proton beams with energy between 30 and 70 MeV and current between $[250 - 500]\mu A$: the facility has been designed in order to serve at the same time two experimental areas (Figure.1.1). The dual-exits SPES cyclotron enables the irradiation of two target at the same time, sharing a total proton beam current of 0.750 mA, the first beam at $200\mu A$ and 40 MeV is needed for the production of radioactive beams (RIBs) and the second beam up to $500\mu A$ and 70 MeV will be devoted to application as radioisotopes production for medical applications (LARAMED project) and high intensity neutron beams generation for material research (NEPIR project)¹. The production of exotic nuclei is based on ISOL technique² that provides low energy secondary beams that are selected by a high resolution mass spectrometer, then ionized by a technique that increase the charge state of ions, called breeding process, and finally reaccelerated by the ALPI machine operating at LNL. The primary beam is provided by the 70p cyclotron at LNL: a 4 straight sector machine that operates at a magnetic field of 1.6 T. The cyclotron accelerates H^- ions that are extracted by stripping process³ to get the proton beams: the magnetic fields deflects the positive proton in the opposite direction from the negative H^- ion and so ejects it from the magnet. The total extraction radius is about 1300 mm and the total weight is 160 tonnes. The 70 MeV radio-frequency system consist of two independent triangular structured cavities separated and shielded at the center design providing a

¹<https://web.infn.it/spes/index.php/characteristics/documents>

²Isotope Separation On-Line. The ISOL method has been associated with thick targets in which the reaction products are thermalised in the target itself and diffuse out to an ion source for further acceleration and separation[12]. For RIBs production the target at the SPES building will be 7 disks of Uranium Carbide compound

³In the extraction process the negative hydrogen ion beam passes a thin carbon foil (e.g. pyrolytic graphite, typically 50 to 200 $\mu g/cm^2$), which strips off the electrons[13].

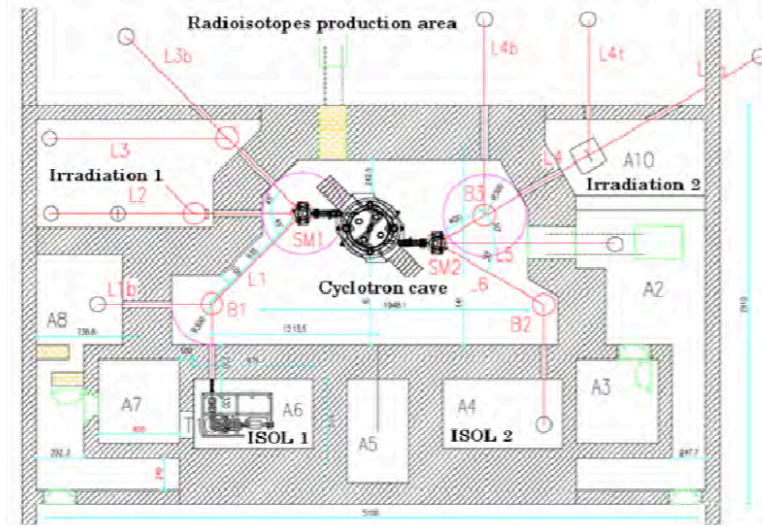


Figure 1.1: Partial layout of SPES. Proton beam distribution

symmetrical electrode (Dee) voltage distribution for optimum beam centering (Figure 1.3⁴): the defocusing effect due to a magnetic field that slightly increase with the radius is compensated through the azimuthal variation of the field.[8].

1.1.1 Radiation fields associated with particle accelerators

Two distinct radiation fields are associated with particle accelerators: the "prompt" radiation, due to accelerator operation, that disappears once it's turned off and the one evaluated in this work, the "remanent" radiation. This "remanent" field, in fact, persists after the accelerator shutdown and it's caused by the radioactivity induced in the accelerator structures.

The cyclotron used to provide the primary beam to SPES project, accelerates proton beams up to 70 MeV that interact both with materials of choice, depending on the application, and with the cyclotron components and its surrounding areas.

The induced activation, in this work, has been considered due to the 70 MeV proton beam during the cyclotron commissioning tests:

1. in H^- beam injection tests, the beam itself has been stopped inside the cyclotron vacuum chamber with a copper interceptor cooled by a water cooling system: in this work has been analyzed and estimated the activation of a sample of this **water**;
2. in proton beam extraction tests, the proton beam exiting the cyclotron has been directed on thick copper and silver targets: in this work the activation of a **copper target** (from now on called beam dump BD) by irradiation of the proton beam at 70 MeV and $1\mu A$ has been measured and guessed;

⁴ Picture from <http://jolisfukyu.tokai-sc.jaea.go.jp/fukyu/tayu/ACT97E/03/0303.htm>

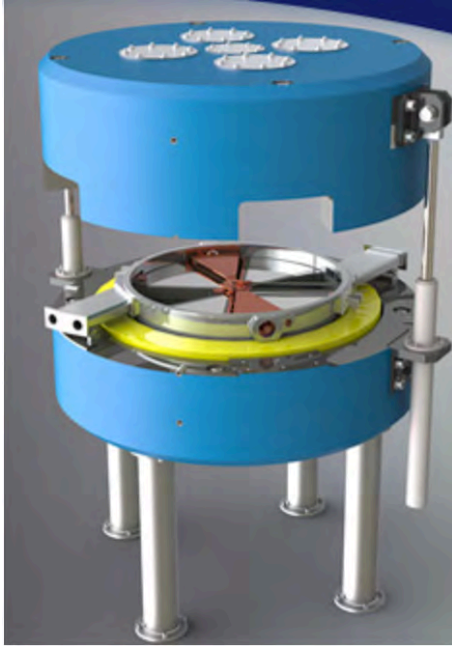


Figure 1.2: Layout of the cyclotron. The upper part of the cyclotron is lifted to allow the view its inside structure, picture from <http://www.bestcyclotron.com>

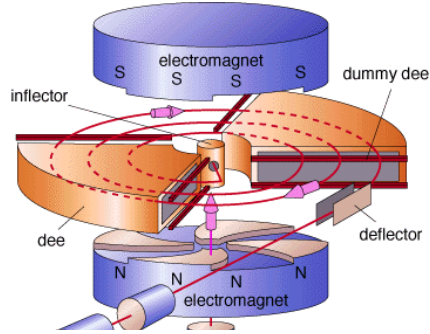


Figure 1.3: Summary scheme of the cyclotron functioning

3. in the last commissioning tests the proton beam (from $100\mu A$ to $500\mu A$) has irradiated a copper target, shielded with lead and polyethylene, inside the irradiation bunker that will host the future SPES project uranium compound target; during the irradiation the activation of some environmental matrices has been studied: in this the activation of a **dust** sample near the proton beam line pipe has been discussed.

The average current of the proton beam used is of $1\mu A$, for the copper beam dump and the cooling water, and of $100\mu A$ for the dust sample.

1.2 Gamma Detectors

A coaxial HPGe at liquid nitrogen temperature with low background shielding, that works at a negative HV=-4000 Vdc and has a relative efficiency of 37.6% at 1332 keV ⁵, has been used to identify radionuclides in water and dust samples, while a portable one was used for the beam dump activation analysis. The efficiency and energy calibration as well as the radioisotopes identification and activities computation were made using

⁵ ^{60}Co

the GenieTM 2000 Spectroscopy Software. This software acquiring and analyzing spectra from Multichannel Analyzers, produces qualitative and quantitative gamma analysis of the samples. The Calibration Software is also capable to generate the necessary efficiency curves combining the detector characterization produced by the MCNP modeling code, mathematical geometry templates, and a few physical sample parameters.

Chapter 2

Prediction of induced activity in target and dumps

2.0.1 Hadron Cascades

At low energies¹ (from few MeV to below 50 MeV) protons entering the target nucleus are capable of inducing various types of nuclear reactions, such as (p, n) , (p, np) , $(p, 2n)$, (p, α) and so on, with the progressive increase of the threshold energy. At higher energy of the incident protons, the possible types of nuclear reaction increase and so does the number of radionuclides produced; one of the most important types among these reactions is spallation[9]. Spallation reaction is defined as a "*nonelastic nuclear interaction induced by a high energy particle (> 50 MeV) producing numerous secondary particles*"² or also as "*a type of nuclear reaction in which the high-energy level of incident particles causes the nucleus to eject more than three particles, thus changing both its mass number and its atomic number*"³ and it can be described as a two-step process[10]:

1. a high energy hadron, as e.g a proton, interacts non-elastically with a target nucleus causing an *intra-nuclear cascade* on a time scale $\approx 10^{-22}s$: the incident particle enters the target atom nucleus depositing energy on nucleons; these nucleons begin to travel through the nucleus hitting other nucleons, thus transferring them energy and giving rise to an intra-nuclear cascade of fast nucleons.
2. the fast nucleons of the intra-nuclear cascade can either escape from the nucleus as secondary particles or give up their energy to excite the whole nucleus. As secondaries, e.g. n, p (or π for interacting hadrons with energy above 1GeV), these fast nucleons can themselves, through inelastic interactions with the target nuclei, produce secondaries: they create an *internuclear cascade* and so they place many further individual nuclei into excited states. Then the nuclei placed into excited states by intra-nuclear cascade nucleons, generated either by primary beam protons

¹using the USAEC classifications, p.99 [1].

²Encyclopedia Britannica

³Nuclear Physics Academic Press

or secondaries, de-excite themselves evaporating off⁴ n, p, d, t, α etc. on a time scale $\approx 10^{-18}/10^{-16}s$.

The variety of radionuclides that can be produced increases with the bombarding particle energy and that is due to the exceeding of more reaction thresholds[9]. Then at high energies one can consider approximatively that all the radionuclides with lower mass number than that of the material of the irradiated target can be produced, even though some of them can be neglected due to their short half-life or small cross section. In figure 2.1 there's a schematic example of high energy *hadronic cascade*.

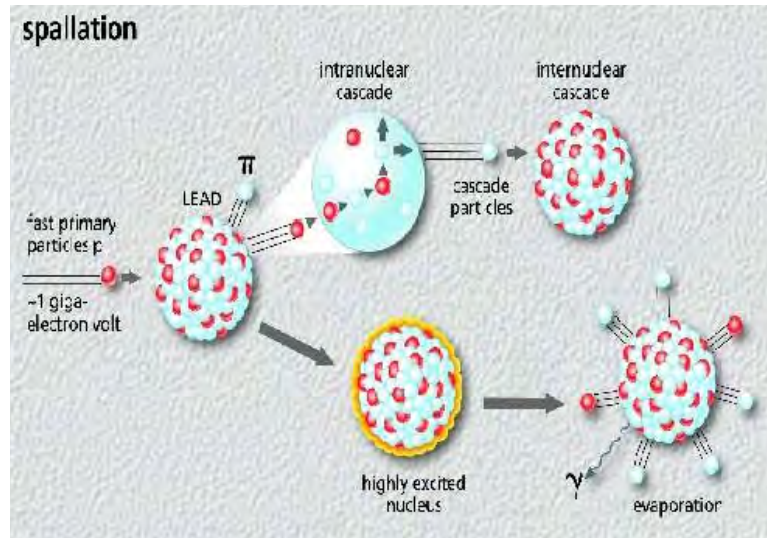


Figure 2.1: High energy particle spallation

At **energies below about 1 GeV** incident protons lose amount of their energy by ionization in the target before interacting with its nuclei. Any secondary charged particle, emitted as a result of an interaction between a proton of less than 1 GeV and a target nucleus, most probably will lose all its energy in the target material by ionization before further nuclear interactions: secondary neutrons from the inter-nuclear cascade are then considered the major component of the secondary radiation[1]. The target nucleus left excited after the initial interaction, de-excites itself evaporating off neutrons of energy of about 2 MeV and gamma rays. As the incident proton energy decreases so does the secondaries energy, determining the rise of the relative importance of evaporation products. Also, fast neutrons with energy below 120 MeV are able to be absorbed by the target more easily than the ones with higher energy, produced by protons with energy $>1\text{GeV}$: they contribute to determine a more complex neutron spectrum. While secondary particles, because of momentum conservation, are strongly concentrated

⁴with evaporation reaction the sequent process is intended: during the intra-nuclear cascade some hadrons deposit their kinetic energy in the target nucleus living it in an excited state; when the nucleus de-excites itself, it emits, or "evaporates" off, principally slow neutrons and gamma rays.

in the forward direction relative to the incident-proton direction, evaporation neutrons are emitted isotropically.

Low energy neutron reactions together with proton spallation interactions are the primary source of induced activity in accelerator: the low energy neutrons are mostly evaporation neutrons slowed down by multiple scattering, and they interact with the accelerator materials principally by the following reactions (n, p) , (n, α) or (n, γ) . The most important reaction among them is (n, γ) , or neutron **radiative capture** that occurs when the neutrons reach thermal energies (about 0.025 eV) due to multiple scattering: *radiative capture* (Figure 2.2⁵) results when low energetic neutrons, as thermal neutrons, are absorbed by a target nucleus, to form the next higher isotope (of mass $A+1$), in an excited state of energy. The new isotope de-excites itself by emitting gamma rays. The neutron is thus lost in this reaction.

Hence **at our 70 MeV proton energy** one can expect proton spallation interactions

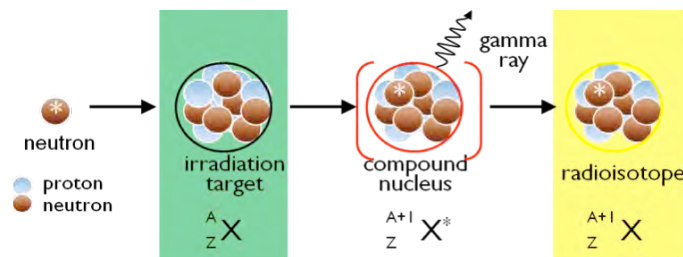


Figure 2.2: Neutron *radiative capture*

in the accelerator materials. Because of the limited range of charged particles at low energies, secondary cascade particles are mostly neutrons of energy of about 25 MeV, that can interact with the target nuclei and produce further radioisotopes. Due to the low proton energy, as previously mentioned, the relevance of evaporation neutrons following spallation interactions increases: their interactions with the accelerator, and among them especially neutron *radiative capture*, can constitute a significant cause of induced activity.

2.1 Induced activity in a copper beam dump by hadron nuclear interactions

To estimate the induced activity in the analyzed samples different analytical methods were applied: they were intended as potential tools that can speedily, unlike computer simulations, give a first rough appreciation of the order of magnitude of the activation and also lead to a closer look to the basics of accelerator activation dynamics.

⁵ Picture from <http://www.keyword-suggestions.com/bmV1dHJvbiBjYXB0dXJl/>

2.1.1 Sullivan Overton approximation

The radionuclides most frequently identified in materials around accelerators, due to high-energy hadron interactions, and with a half-life [10 min;3y], are listed in Table 2.1: the most likely radionuclides produced are those with a mass number from ${}^7\text{Be}$ ⁶ to the mass number of the accelerator material itself [1]. The number of radionuclides generated by proton interactions can appear quite large under the assumption that all radionuclides lighter than the irradiated material can be produced: in copper it results that about 60 radionuclides can be generated. The production of most of them is however negligible: the copper activity has been attributed only to those that are both gamma emitters and with a half-life greater than five hours: with a cooling time greater than one day, in the copper beam dump, the activities relative to isotopes with a half-life smaller become very slight or undetectable. Therefore, going through the isotopic table⁷, it can be inferred that there's a chance of about 13% that the isotope produced by spallation will be radioactive and with a half-life in the range [5h; 3y]⁸. The **Sullivan and Overton (S.O.) approximation**[3], useful in the chosen half-life range, suggests that the probability of an isotope of having a given half-life, τ , is inversely proportional to the half-life itself. The number of isotopes of a half-life τ , in a range from τ to $\tau + \delta\tau$, $N\delta\tau$, can be considered therefore proportional to $\delta\tau/\tau$. Then, the probability that a nuclear isotope, generated by a single interaction between a medium energetic hadron and a target nucleus, will have a half-life τ is⁹:

$$P(\tau)\delta\tau = 0.13 \frac{N(\tau)\delta\tau}{\int_{\tau_2}^{\tau_1} N(\tau)\delta\tau} \quad (2.1)$$

where $\tau_1=3\text{y}$ and $\tau_2=5\text{h}$. Using then S.O. approximation, $P(\tau)\delta\tau \propto \frac{\delta\tau}{\tau}$, $N\delta\tau=\delta\tau/\tau$,

$$P(\tau)\delta\tau = 0.13 \frac{\delta\tau}{\tau \ln(\tau_1/\tau_2)} = 0.015 \cdot \delta\tau/\tau. \quad (2.2)$$

The activation equation and S.O. approximation

Supposing a steady irradiation of a material by a spatially uniform flux density of hadrons, the simplest activation, per unit volume, equation for a single radionuclide of decay half-life τ is

$$a(t) = N\sigma\phi(1 - e^{-0.693t/\tau})(e^{-0.693t/\tau}), (Bq/cm^3) \quad (2.3)$$

where $N\sigma\phi$ is the rate gain of the radionuclide through the production reaction considered, with N , the the number density of target atoms [cm^{-3}], ϕ the flux density [$cm^{-2}s^{-1}$] of the incident particles and σ the production cross section [cm^2]. $N\sigma$ is an index of the collision probability during the passage of hadrons through the medium. The first factor

⁶the proton energy threshold to produce this Berillium isotope is around 100MeV

⁷<https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.htm>

⁸p.3 [11]

⁹p.103 [3]

| Material | Isotope | Threshold (MeV) | τ |
|-------------------|---------------------------|-----------------|---------|
| Plastics and Oils | 7Be | 2 | 53.22 d |
| Al | As above plus ${}^{22}Na$ | 30 | 2.60 y |
| Iron | As above plus | | |
| | ${}^{44m}Sc$ | | 2.44d |
| | ${}^{46}Sc$ | | 83.8d |
| | ${}^{48}Sc$ | | 1.82d |
| | ${}^{48}V$ | 20 | 15.97d |
| | ${}^{51}Cr$ | 30 | 27.7d |
| | ${}^{52}Mn$ | 20 | 5.59d |
| | ${}^{54}Mn$ | | 312.1d |
| | ${}^{55}Fe$ | | 2.74y |
| | ${}^{59}Fe$ | | 44.5d |
| | ${}^{56}Co$ | 5 | 77.2d |
| | ${}^{57}Co$ | 30 | 271.7d |
| | ${}^{58}Co$ | 30 | 70.9d |
| Copper | As above plus | | |
| | ${}^{65}Zn$ | | 243.7d |

Table 2.1: Most common radioisotopes with half-life in $[\tau_2; \tau_1]$ found in irradiated materials around accelerators [9].

of eq. (2.3) describes the activity build up during the irradiation time T ; the second one instead represents its decay as a function of the cooling time, t .

If the copper target activity is due to 70 MeV hadron spallation interactions, because of the complexity of the process, it's difficult to promptly and precisely guess which radionuclides are produced: S.O. approximation comes in hand to calculate a rough estimate of the copper BD activation.

If one considers the previous activity equation (2.3), modifies the flux so that there's only one proton interacting with a target nucleus and also changes the index of collision probability $N\sigma$ with the probability (previously computed in equation (2.2)) that a nuclear isotope generated by the interaction will have a half-life τ , then the specific activation from one spallation interaction per second in copper for an isotope of half-life in the range $[\tau; \tau + \delta\tau]$ is expressed by the following equation:

$$S_{1sp}(\tau)\delta\tau = P(\tau)(1 - e^{-0.693T/\tau})(e^{-0.693t/\tau})\delta\tau. \quad (2.4)$$

The activity induced by a single radionuclide, produced by a proton spallation interaction in copper, having a τ , half-life, is hence computed under the following assumptions: the build up and decay of the radionuclide is described as in equation (2.3), $(1 - e^{-0.693T/\tau})(e^{-0.693t/\tau})$, while its probability of having the τ half-life in the range $[\tau; \tau + \delta\tau]$, is given by equation (2.3).

An estimation of the activity due to one spallation interaction per second, can be therefore obtained by integrating eq. (2.4) over all of τ values in the interval of interest $[\tau_2; \tau_1]$: that means that the total activity from one spallation interaction is therefore computed considering all the possible isotopes activated, with τ between 5h and 3y, weighted by their probability production in it, $P(\tau)$. The induced activity in the copper target from one medium energy hadron spallation interaction per second therefore results

$$S_{1sp} = 0.015 \int_{\tau_2}^{\tau_1} \frac{(1 - e^{-0.693T/\tau})(e^{-0.693t/\tau})}{\tau} \delta\tau = 0.015 \ln\left[\frac{T+t}{t}\right], (Bq). \quad (2.5)$$

2.1.2 Radioactivity in copper beam dump from the 70 MeV beam proton nuclear interactions

Proton spallation reactions and neutron inelastic interactions can be considered the main causes of induced radioactivity in the copper beam dump analyzed. The three activation sources discussed below are (1) spallation interactions from the primary proton beam, (2) inelastic interactions from secondary *cascade* neutrons and (3) neutron *radiative capture* reactions. The activation due to secondary proton, as before mentioned, is neglected because of their important ionization energy loss before any nuclear interaction.

The number of spallation interactions per second in a copper target of thickness¹⁰ $1g.cm^{-2}$, bombarded by a medium energy proton (70 MeV), is $\frac{1}{\lambda}$, where λ is the interaction mean free path in copper. The mean free path of proton of 70 MeV in copper is related to the inelastic nuclear cross section σ , by

$$\lambda = \frac{A}{N_A \sigma}, (g.cm^{-2}) \quad (2.6)$$

where N_A is the Avogadro's number ($6.02 \cdot 10^{23}$ atoms per mole), A is the atomic weight of the material and σ is the nuclear interaction cross section in cm^2 .

The nuclear interaction cross section for 70 MeV protons in copper is considered $\sigma = 2 \cdot 10^{-25} cm^2$ ¹¹[?] and main free path results therefore $\lambda = 527.61g/cm^2$.

Hence, the induced activity per $g.cm^{-2}$ in a copper target due to the primary proton beam of $\phi = 6.24 \cdot 10^{12}(proton/s)$,

$$S = \frac{1}{\lambda} \phi S_{1sp} = \frac{1}{\lambda} \phi \cdot 0.015 \ln\left[\frac{T+t}{t}\right], (Bq) \quad (2.7)$$

where S_{1sp} is the activity per single interaction per second calculated in (2.5) with $t=1800$ (min) the cooling time, $T=150$ (min), the irradiation time. In addition, the fact that not all the beam protons will interact before coming to rest, due to ionization energy loss, has to be taken into consideration: their approximate mean interacting fraction is about

$$f = 1 - e^{-\frac{R}{\lambda}} \quad (2.8)$$

¹⁰The interaction depth unit

¹¹is the compressive cross section of all possible radionuclides produced by proton spallation on copper, under our criterions, previously mentioned

where $R = 6.314g.cm^{-2}$, the 70 MeV proton range in the target material. Eq. (2.7), multiplied by the copper density and mean interaction fraction, hence results

$$S_p = S \cdot f \cdot \rho = 1.56, (MBq.cm^{-1}) \quad (2.9)$$

effective activity per cm induced by proton spallation interactions in the copper beam dump. Considering then a proton penetration depth in the copper BD of about 0.8 cm, the total estimate of the activity induced by proton interactions in the copper BD is $S_{pt} = 1.25(MBq)$.

Gamma dose rate from proton induced activity

The average gamma dose rate at 1 m, from the mixture of spallation produced isotopes, of the previously computed activity S_{pt} , is computed through the following equation:

$$D = k_s S_{pt}, (Sv.h^{-1}) \quad (2.10)$$

where k_s is the conversion factor, the gamma dose rate constant, that converts radioactivity levels into dose rate and defines the radiological importance of the isotopes concerned. For isotopes emitting several different energy photons per disintegration, the k_s factor is the sum of the dose rate at 1 m from all the gamma rays per disintegration per second¹². In copper, it can be estimated that about 1.6 photons are emitted per decay of a spallation produced isotope¹³. If then, k has a value of $120 fSv.Bq^{-1}.h^{-1}$ at 1 m, for isotopes emitting a single gamma photon of $0.8MeV$ per decay, hence, the effective average value of k_s , for a 1.6 photon emission, becomes $k_s = 192 fSv.h^{-1}.Bq^{-1}$ at 1 m.

The activity in the copper beam dump, previously calculated, $S_{pt} = 1.25 MBq$: the dose rate from proton spallation induced activity on the copper dump is then equal to: $D = 2.40 \cdot 10^{-4} mSv.h^{-1}$, at 1 m from the mixture of spallation produced isotopes.

2.1.3 Radioactivity in copper targets from secondary neutron nuclear reactions

As mentioned at the beginning of paragraph (2.1.2), secondary particles, especially cascade neutrons, can themselves interact with the copper beam dump.

If the hadron cascade is composed primarily by neutrons, to calculate the activation of the copper target, the following estimates are necessary: the flux and the mean energy of the secondary neutron beam. From the last, the nuclear inelastic interaction cross section of the neutrons in the copper dump can be found in literature.

The total neutron emission per proton interaction, or **multiplicity**¹⁴ $Q=2.2$ neutrons per incident proton on the copper BD where $Q_s = 0.4$, is the multiplicity of the cascade

¹²p.96[3]

¹³ In our case in fact 7% of the decays the daughter nucleus of the radionuclides generated in the copper BD, is likely to be active: the effective average photon emission per decay of an induced radioactive isotope in the BD is near 1.6

¹⁴p.136 [1]

neutrons. The average energy of the secondaries is determined assuming that protons of energy less than 1 GeV incident on a thick target, lose on average 20% of their energy by ionization,

$$E_n = 0.8 \frac{E}{Q} = 25, (MeV). \quad (2.11)$$

Secondaries, or cascade, neutrons at 25 MeV interact with the copper nuclei giving rise especially to radionuclides with atomic mass number close to that of the target bombarded[1]. $S_{1sp.n}$, the activity induced in copper by a single neutron nuclear interaction with the beam dump, is obtained taking into consideration that, going through the isotopic table, there's an approximative chance of 0.98% that the isotopes produced by neutron inelastic interaction with copper will be a radioactive gamma emitter with a half-life in the range previously chosen $[\tau_2, \tau_1]$: the only radionuclide that fulfills this requests is ^{64}Cu , with a half-life of 12.7h. The production cross section of the isotope is [5] $\sigma = 295 \cdot 10^{-27} cm^2$ from which, through (2.6), $\lambda_n = 357 g/cm^2$.

The effective multiplicity of secondaries approaches to fQ_s , with, f, factor discussed in the prior paragraph. It's now possible to estimate the activity of the copper BD irradiated with a flux of secondary neutrons equal to $\Phi_n = fQ_s\phi$ (neutrons/s), where ϕ indicates original proton flux previously used in eq. (2.7). The secondary neutrons induced activity is considered due only to ^{64}Cu isotope, with T and t the same as those used in eq.(2.7), therefore it results:

$$S_n = \frac{1}{\lambda_n} \Phi_n \cdot S_{1sp.n} = 0.0098 \cdot \frac{1}{\lambda_n} \Phi_n (1 - e^{-0.693 \frac{T}{\tau}}) \cdot e^{-0.693 \frac{t}{\tau}} = 1.98 \cdot 10^4 (Bq.g^{-1}.cm^2). \quad (2.12)$$

The activity per cm, obtained multiplying (2.12) by the copper density $\rho = 8.96 g/cm^3$, is $S_{n\rho} = 1.76 \cdot 10^5 Bq.cm^{-1}$; this activity has then been multiplied by the effective proton penetration depth, 0.8 cm, $S_{ntot} = 1.41 \cdot 10^5 Bq$. Using $k_\gamma = 3.514 \cdot 10^{-11} mSv/(h.Bq)$ at 1m distance and the cascade neutron induced activity S_{ntot} in eq. (2.10), the dose rate on the copper beam dump is then equal to $D_n = 4.96 \cdot 10^{-6} mSv.h^{-1}$ at 1 m.

2.1.4 Radioactivity in copper targets from neutron *radiation capture*

As previously mentioned, the energy distribution of neutrons generated in the copper BD by irradiation of a 70 MeV proton beam is divided mostly into two groups: low energy evaporation neutrons emitted isotropically and more energetic cascade neutrons, with an angular distribution peaked in the forward direction. Evaporation neutrons, because of their lower energy, are most likely slowed down to thermal energy through multiple scattering and then get captured by the target nuclei in the reaction (n, γ) . The estimate **number of evaporation neutrons** produced per incident 70 MeV proton per inelastic collision is about 1.8¹⁵. However, evaporation neutrons don't lose significant energy through the copper BD thickness: they instead will contribute to the activation of the BD surrounding materials as concrete and aluminium.

¹⁵[3] page 136 Fig. 3.32

Dose rate from the copper beam dump

The dose rates from isotopes produced in the BD copper interaction with, respectively, primary protons and secondary neutrons

| D | $mSv.h^{-1}$ |
|-----------|----------------------|
| D_p | $2.40 \cdot 10^{-4}$ |
| D_n | $4.96 \cdot 10^{-6}$ |
| D_{tot} | $2.45 \cdot 10^{-4}$ |

Table 2.2: Copper beam dump dose rate

2.2 Radioactivity induced in the cyclotron cooling water

The rough estimate of the induced activity in the cyclotron cooling water has been made under the following approximations of the system geometry: the cooling water DDT follows the path of a pipe that originates from a 8l capacity water tank; the pipe continues for about 3m and then coils itself around the copper beam interceptor sides (for about 24 cm) where the direct beam line doesn't come across; it progresses, moving away from the interceptor, for other 3 meters until it reaches again the tank where the water is completely mixed and put again in circulation. As mentioned the primary H^- beam on the copper interceptor, through spallation interactions, gives rise to cascade hadrons, fast protons and neutrons¹⁶. Due to the secondary proton energy loss by ionization, the cascade neutrons are the major component of the secondary radiation. Also, evaporation neutrons and protons are most likely stopped respectively by multiple scattering and ionization, before interacting with water nuclei: the cooling water can be considered irradiated mostly by the cascade neutrons. It had to be taken in mind that even though the secondary radiation is predominantly forward directed, this predominance increases with increasing energy, in our case in fact the forward fluence $\phi(0)$ is about 7.63 times the fluence at 90 degrees from the beam forward direction $\phi(90)$, both computed from equation (2.15).

The irradiation model used is the Laminar Flow Model [6] where the fluid passes at a uniform speed through the irradiation region without turbulence. Considering that the water pipe has radius of 0.5 cm, the total volume of water cycled trough the all pipe is $V_{irr} = 0.49l$ and the flow rate in the pipe is $Q = \frac{5l}{min}$, the water flux trough the pipe is kept constant: v , the linear velocity of the fluid in the tunnel or pipe is therefore equal to

$$v = \frac{QL}{V_{irr}} = 106 \left(\frac{cm}{s} \right). \quad (2.13)$$

with $L=624cm$, the total water path length. The irradiation time, near the copper interceptor, is $t_{irr} = \frac{l_{irr}}{v} = 0.23s$, with $l_{irr} = 24cm$ the water irradiation path length, and

¹⁶the interaction of H^- ions has been considered analogue to that of protons, neglecting Barkas effects

the cooling time after irradiation is $t_{dec} = \frac{l_{col}}{v} = 2.83s$, with $l_{col} = 3m$ the water cooling path length¹⁷. In the first cycle through the pipe, the water circulating is irradiated for 0.23 s and it's activity decays for 2.83 s before reaching the water tank (as describes the first term of equation (2.14)); in the second cycle the volume of water circulating is taken from the tank where the activity resulting from the first cycle has been diluted in the 8l tank capacity: it decays for further 2.83 s, is then irradiated for 0.23 s and decays again for 2.8s before reaching the tank where it dilutes itself newly. If the total copper interceptor irradiation time is of 204 min, after the first cycle, characterized only by one irradiation and one decay time, around 2082 cycles follow characterized by one irradiation and two decay times (second term of equation (2.14)).

If the water after the irradiation time is taken directly from the water tank, the activation is:

$$A = N\sigma\Phi_n(90) \cdot \left[\frac{1}{8} e^{-0.693 \frac{t_{dec}}{\tau}} \cdot (1 - e^{-0.693 \frac{t_{irr}}{\tau}}) + \frac{2082}{8} \cdot e^{-0.693 \frac{t_{dec}}{\tau}} \cdot (1 - e^{-0.693 \frac{t_{irr}}{\tau}}) \cdot e^{-0.693 \frac{t_{dec}}{\tau}} \right] (Bq/cm^3) \quad (2.14)$$

where $\Phi_n(90)$ is the secondary neutron flux density at 90 degrees from the H^- beam line in $cm^{-2} \cdot s^{-1}$, N is the number density of the target atoms in cm^{-3} and σ the production cross section of the activated isotope.

Cascade neutron flux density calculation

Cascade neutron fluence on the water sample is obtained from the cascade neutron properties, on copper, previously calculated: the multiplicity, Q , the secondary neutron emission per proton interaction from eq. (2.9), $E_{sec} = 25MeV$, the average energy of the secondaries and $f = 0.01$, the mean proton fraction interacting with the copper layer from eq. (2.8). Combining fQ with the angular distribution of secondaries at high proton energies although corrected for interaction probability and ionization losses¹⁸, the angular distribution of the fluence of secondaries per low energy proton incident on a copper target is

$$\phi(\theta) = \frac{5000 \cdot f}{(\theta + 40/\sqrt{E})^2}, neutrons.m^{-2} \quad (2.15)$$

at 1m and an angle θ to the incident H^- direction of energy E in GeV. With $\theta = 90^\circ$ and $E=70$ MeV, f previously calculated¹⁹, then $\phi(90) = 6.38 \cdot 10^5$, ($neutrons.cm^{-2}$) and $\Phi_n(90) = \phi(90) \cdot \phi_{H^-}$, with ϕ_{H^-} , flux of H^- per second (considered equal to the extracted proton flux).

¹⁷the cooling path length for the water is equal before entering the irradiation zone, from the tank, and exiting it going toward the tank

¹⁸p.48/49[3]

¹⁹See footnote 15.

Radioisotopes production

Due to their similar mass, hydrogens and neutrons are most likely to elastically interact: the induced activity is produced by neutron nuclear reaction with the oxygen isotope ^{16}O (present in 99.6% of water molecules). The most likely production reaction between secondary neutrons and ^{16}O in water are $^{16}\text{O}(n, sp)^7\text{Be}$, $^{16}\text{O}(n, sp)^{13}\text{N}$, $^{16}\text{O}(n, sp)^{11}\text{C}$ and $^{16}\text{O}(n, 2n)^{15}\text{O}$: they in fact are the only gamma emitters with atomic mass lower than $A_{^{16}\text{O}}$ and greater than A_H and half-life between 2 minutes and 3 years. However the Berillium isotope, has significant spallation cross section only interacting hadrons with energy above 1GeV, therefore the expected induced activity is considered due only to ^{13}N , ^{11}C and ^{15}O , also usually the most common radionuclides produced in hadron spallation on ^{16}O .

The number of water molecules (in mol) is equal to the number of ^{16}O atoms multiplied by 0.996, therefore the isotope weight in grams in the water sample of 1l is equal to

$$^{16}\text{O}_{weight} = H_2\text{O}(mol) \cdot 0.996 \cdot A_{^{16}\text{O}} = \frac{1kgH_2O}{A_{H_2O}} \cdot 0.996 \cdot 15.99 g \cdot mol^{-1} = 884.24 g \quad (2.16)$$

Therefore to calculate N, the oxygen isotope density can be derived from $N = \frac{\rho N_A}{A_{^{16}\text{O}}}$ with $\rho = \frac{884.24 g}{1000 cm^3}$.

The specific activity of the produced radioisotopes has been derived trough equation (2.14) :

| Isotope | $\sigma [cm^2]$ | $\tau [s]$ | Act. [$Bq \cdot cm^{-3}$] |
|-----------------|--------------------|------------|-----------------------------|
| ^{15}O | $4 \cdot 10^{-26}$ | 121 | $3.71 \cdot 10^3$ |
| ^{13}N | $1 \cdot 10^{-26}$ | 598 | $1.91 \cdot 10^2$ |
| ^{11}C | $2 \cdot 10^{-26}$ | 1218 | $1.88 \cdot 10^2$ |

The 1l sample has been cooled for about another 2 minutes before its activity detection, so the activities have to be multiplied by the exponential factor $e^{-0.693 \cdot \frac{2min}{\tau}}$. In the following table²⁰ are listed the estimates of the activity after the sample cooled for 2 minutes in Bq/l and their equivalent dose.

| Isotope | Act. [$MBq \cdot l^{-1}$] | Dose rate [$mSv \cdot h^{-1}$] |
|-----------------|-----------------------------|----------------------------------|
| ^{15}O | $2.94 \cdot 10^{-2}$ | $3.55 \cdot 10^{-10}$ |
| ^{13}N | 0.76 | $3.18 \cdot 10^{-11}$ |
| ^{11}C | 1.27 | $3.36 \cdot 10^{-11}$ |

$D_{tot} = 4.20 \cdot 10^{-10} mSv/h$ at 1 m distance.

²⁰p.138 [1], where all β^+ isotopes are assumed also to emit 2x0.511 MeV photons, due to positron, electron annihilation

2.3 Induced activity in dust near the beam line pipe in the irradiation cave

Internal contamination by inhalation of radioactive dust produced by activated parts of the cyclotron machine, causes some radiation hazard for maintenance staff, additional to the external exposure from activated cyclotron parts. The production of radioactive nuclides can be explained considering that the dust activation is due to the particle bombardment of the tungsten-made septum of the beam duct²¹: the intense particle beam, bombarding the tungsten septum, gives rise to radionuclides as $^{183,184}\text{Re}$, ^{185}Os and ^{182}Ta that evaporate from the septum hot spot and deposit on the inner surface of the cyclotron chamber. The introduction of air into the chamber leads these nuclides to the dust in the cyclotron chambers and so they're often find by smear test even in the remote place of the cyclotron machine. In the dust it has been also revealed the presence of ^{65}Zn due to heating and subsequent evaporation of copper parts of the cyclotron and co-isotopes, among which ^{60}Co produced by (n, α) or (n, p) reactions, and ^{22}Na from $(n, 2n)$ reactions in sodium. The presence of ^7Be is instead attributed to the activation of the graphite cover used to protect the edges of the dees near the ion source from melting.

In our case however the dust is not an activation and exploitation product from the machine parts: for activation tests instead it has been collected, inside a cuvette, in the cyclotron irradiation cave and placed near the beam line pipe, in close proximity with the copper target irradiated. The copper target irradiated with a $100\mu\text{A}$ and 70 MeV proton beam, is shielded with lead and polyethylene: the secondary radiation fast neutrons, caused by the proton beam interaction with the copper itself, are then slowed down by the target shielding through multiple scattering until they reach almost thermal energies. Thermalized neutrons exiting the shielding can hence be considered as the primary constituent of the radiation interacting with the dust sample.

The sample collected in the irradiation cave is most likely composed by concrete from the cave walls and small traces of soil and earth from the cave ground. **Sodium** in concrete, **manganese** in earth and concrete and natural **potassium** in soil have a significant thermal activation cross section along with **cobalt**, **caesium** and **europium** in earth and concrete²². Some of the reactions products that can therefore be expected, in ascending order of thermal cross section, are: $^{23}\text{Na}(n, \gamma)^{24}\text{Na}$, $^{41}\text{K}(n, \gamma)^{42}\text{K}$, $^{55}\text{Mn}(n, \gamma)^{56}\text{Mn}$, $^{59}\text{Co}(n, \gamma)^{60}\text{Co}$, $^{133}\text{Cs}(n, \gamma)^{134}\text{Cs}$, $^{151}\text{Eu}(n, \gamma)^{152}\text{Eu}$.

²¹[7]

²²p.120 [3]

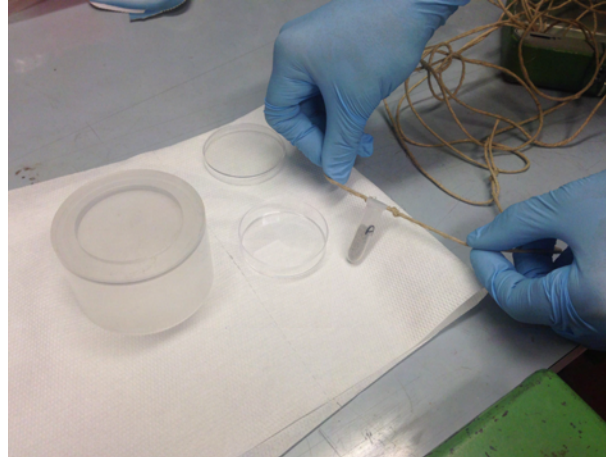


Figure 2.3: Dust sample collection after irradiation



Figure 2.4: Dust sample inside the HPGe gamma detector well.

Chapter 3

Measurement campaign and collected data

Copper beam dump

The radioisotopes listed were revealed in the copper beam dump with spectroscopic techniques through a portable HPGe-based gamma detector (paragraph 1.2) after about 2 and a half hour irradiation and a day and a half of cooling time¹

| Isotope | τ | Act.[Bq] | Dose rate[mSv.h ⁻¹] |
|------------------|--------|----------------|---------------------------------|
| ²² Na | 2.6 y | 5000 ± 1000 | 1.80 · 10 ⁻⁶ |
| ²⁴ Na | 15 h | 4480 ± 1000 | 2.29 · 10 ⁻⁶ |
| ⁴⁶ Sc | 83 d | 2580 ± 640 | 8.14 · 10 ⁻⁷ |
| ⁵¹ Cr | 27 d | 126000 ± 37000 | 7.96 · 10 ⁻⁷ |
| ⁵⁴ Mn | 312 d | 91000 ± 5000 | 1.26 · 10 ⁻⁵ |
| ⁵⁶ Co | 77 d | 33000 ± 1400 | 1.70 · 10 ⁻⁵ |
| ⁵⁸ Co | 70 d | 170000 ± 9300 | 2.81 · 10 ⁻⁵ |
| ⁶⁵ Zn | 243 d | 62000 ± 3300 | 5.44 · 10 ⁻⁶ |

Table 3.1: Copper beam dump activities and dose rates revealed

$D_{tot} = 6.88 \cdot 10^{-5} mSv/h$ at 1 m distance. The copper BD dose rate analytically derived ($2.45 \cdot 10^{-4} mSv/h$), besides parameters approximation and mostly due to the fact is based on the sum of all the probable radioisotopes (gamma emitters, produced in the half-life range of choice [5h;3y]), appears overestimated by a factor of 3.6.

The radioisotopes detected have a half-life and atomic mass in the ranges expected by the 70 MeV proton spallation on copper: τ in the range [5h; 3y] and atomic mass between

¹dose rate values are obtained multiplying the activity of each radionuclide with their specific gamma rays dose constants[9]

[7;65]u. Radioisotopes that don't decay significantly during the cooling time and have both high production cross section (c.s [4-40]mb) and radiation intensity are indeed more likely to be detected: among them in the table above one can find ^{65}Zn , ^{58}Co , ^{56}Co , ^{54}Mn and (with lower c.s. 0.4 mb) ^{46}Sc . Otherwise low gamma intensity emitters (and even more with half life <24h) can be easily lost in the background spectrum (e.g. in our case ^{64}Cu with $\sigma = 40\text{mb}$ and $\tau = 15\text{h}$ and ^{70}Co with $\tau = 271\text{d}$ and $\sigma = 40\text{mb}$). Low intensity positron emitters can't also be directly identified due to their simultaneous contribution to the escape peaks (annihilation radiation).

Due to the fact that this was not a low background gamma activity measurement the activity of some radioisotopes can be attributed, in addition to proton interaction on copper, even to the activation of some materials near the BD: e.g. the presence in the spectrum of sodium isotopes can be explained also through secondary neutron interactions with aluminium, such as $^{27}\text{Al}(n, x)\text{Na}$ and $^{27}\text{Al}(n, x)^{24}\text{Na}$ and that of chromium by the secondary neutron activation of iron e.g. $^{24}\text{Fe}(n, x)^{51}\text{Cr}^2$.

Cooling water

A sample of the water circulating in the copper beam interceptor cooling system has been taken right after about 3 and a half hours of proton beam irradiation of the beam interceptor. The cooling system under investigation is the one dedicated to the copper beam interceptor inside the cyclotron (in the first commissioning phases, some tests of beam acceleration without extraction have been done, so the beam had to be stopped inside the cyclotron vacuum chamber). The radioactivity has been evaluated about 2 minutes after the end of irradiation. The activity of some positron emitters, that contribute to the same escape peaks, has been measured $1.98 \pm 0.7 \text{ MBq/l}$. The radioisotopes to which has been attributed this activity are mostly ^{13}N and ^{11}C , due to their decay time and production probability by neutron spallation on ^{16}O in the water sample: other beta emitters are in fact likely to be produced (e.g. ^{14}O , ^{15}O and ^{10}C) but they're not detectable at the time of the measurement due to their short half-life compared to the sample cooling time.

| Isotope | τ | Act. [MBq.l^{-1}] | Dose rate [$\text{mSv.h}^{-1}.\text{l}^{-1}$] |
|-----------------|---------|------------------------------|---|
| ^{13}N | 10 min | 1.98 ± 0.7 | $3.82 \cdot 10^{-10}$ |
| ^{11}C | 20 min | | |
| ^{15}O | 2.1 min | | |

Table 3.2: Cooling water activities measured and dose rate derived

$$D_{tot} = 3.82 \cdot 10^{-10} \text{ mSv/h at 1 m distance.}$$

Hence, even for the water sample the induced activity rough appreciation by the use of analytical methods, although based on approximate parameters, results then able to give

²or $^{24}\text{Fe}(n, x)^{50}\text{Cr}$, and through radiative capture $^{50}\text{Cr}(n, \gamma)^{51}\text{Cr}$

a dose rate estimate with the same order of magnitude of that derived from experimental data ($4.20 \cdot 10^{-10} \text{ mSv/h}$).

Cyclotron cave dust

A sample of the dust collected in the irradiation cave has been measured right after 2 hours of irradiation with $100\mu\text{A}$ of a copper beam dump shielded with lead and polyethylene. Despite the limited duration of the irradiation, it has been possible to identify through a HPGe gamma detector some radionuclides of medium half-life listed below (the specified activities refer on the entire dust sample of few grams weight):

| Isotope | τ | Act.[Bq.] | Dose rate[mSv.h ⁻¹] |
|------------------|--------|--------------|---------------------------------|
| ²⁴ Na | 15 h | 144 ± 5 | $7.37 \cdot 10^{-8}$ |
| ⁵⁶ Mn | 2.6 h | 160 ± 4 | $3.96 \cdot 10^{-8}$ |
| ⁴² K | 12.3 h | 16 ± 1.4 | $6.15 \cdot 10^{-10}$ |

Table 3.3: Dust activities and dose rates

$D_{tot} = 1.14 \cdot 10^{-7} \text{ mSv/h}$ at 1 m distance. The isotopes detected, as explained in paragraph (2.3), are characteristic of heart, concrete and soil thermal neutron capture activation.

3.1 Conclusions

The 70 MeV proton beam, at $(1 - 100)\mu\text{A}$, accelerated by the 70p cyclotron used to provide the primary beam to SPES project, causes induced activity in the samples analyzed:

1. the radionuclides detected in the thick copper target, used in proton beam extraction tests, have half-life and atomic mass in the ranges expected by the 70 MeV proton spallation on copper³; although, due to the fact that this was not a low background gamma activity measurement⁴, the presence in the spectrum of some radioisotopes can be attributed, in addition to proton interaction on copper, even to the activation of some materials near the BD (e.g. the sodium isotopes detection can be explained also through secondary neutron interactions with aluminium, such as $^{27}\text{Al}(n, x)\text{Na}$ and $^{27}\text{Al}(n, x)^{24}\text{Na}$ and that of chromium by the secondary neutron activation of iron e.g. $^{24}\text{Fe}(n, x)^{51}\text{Cr}$ ⁵).
2. the water sample comes from the cooling system of a copper interceptor that is used to stop the beam inside the cyclotron vacuum chamber during injection beam tests. In this sample are detectable medium-life beta emitters and their production is compatible with fast neutron spallation on ¹⁶O: the proton beam interacting with

³ τ in the range $[5h; 3y]$ and atomic mass between $[7; 65]\text{u}$

⁴ in fact a portable HPGe gamma detector has been used

⁵ or $^{24}\text{Fe}(n, x)^{50}\text{Cr}$, and through radiative capture $^{50}\text{Cr}(n, \gamma)^{51}\text{Cr}$

the copper interceptor generates a secondary neutron radiation that than interacts with the cooling water that acts as a moderator; due to their similar masses, hydrogens and neutrons are most likely to elastically interact, hence the induced activity is produced by neutron nuclear reaction with the oxygen isotope ^{16}O (present in 99.6% of water molecules). In our case the most likely detectable production reactions between secondary neutrons and ^{16}O in water are in fact $^{16}\text{O}(n, sp)^{13}\text{N}$, $^{16}\text{O}(n, sp)^{11}\text{C}$ ⁶.

3. the dust sample, for activation tests, has been collected inside the irradiation cave and then placed near the beam line pipe in proximity to a copper beam dump shielded with lead and polyethylene⁷ : the isotopes detected in the sample are most likely activated by thermal neutron radiative capture. The secondary radiation fast neutrons, caused by the proton beam interaction with the copper itself, are in fact slowed down by the target shielding through multiple scattering until they reach almost thermal energies. Thermalized neutrons exiting the shielding can hence be considered as the primary constituent of the radiation interacting with the dust sample: the sample collected in the irradiation cave is most likely composed by concrete from the cave walls and small traces of soil and earth from the cave ground where sodium in concrete, manganese in earth and concrete and natural potassium in soil have a significant thermal activation cross section.

The dose rates from the activities detected on water and dust samples, after the 70p cyclotron commissioning tests, result not relevant from a radiological point of view: the water equivalent dose rate, $D_w = 0.004 \frac{\mu\text{Sv}}{\text{y}}$, and the dust equivalent dose rate, $D_d = 1 \frac{\mu\text{Sv}}{\text{y}}$, are both less than $10 \frac{\mu\text{Sv}}{\text{y}}$ ($10 \mu\text{Sv}$ is the daily dose rate due to background radiation, that, translated into the probability of biological harm, correspond to a probability of death from a fatal tumor of 5 in a million). Whereas the cooling time expected for the copper thick target (or BD) to reach a dose equal to $10 \frac{\mu\text{Sv}}{\text{y}}$ is about 4 days⁸ in which it will require monitoring.

The analytical methods used, although based on approximate parameters ⁹, were able to give rough estimates of the samples dose rates with the same order of magnitude of the dose rates derived from experimental data.

⁶they are indeed the only gamma emitters (annihilation radiation) with atomic mass lower than $A_{^{16}\text{O}}$ and greater than A_{H} and half-life between 2 minutes and 3 years that are likely to be produced: Berillium isotope, has significant spallation cross section only for interacting hadrons with energy above 1GeV.

⁷inside the irradiation bunker that will host the future SPES target

⁸ $D_{BD} = X \cdot \ln(\frac{t+T}{t}) = 68 \frac{\mu\text{Sv}}{\text{h}}$ for $t=1800\text{min}$, cooling time and $T=150\text{min}$, irradiation time. Then $X=862 \frac{\mu\text{Sv}}{\text{y}}$ and with $D_{BD} = X \cdot \ln(\frac{t+T}{t}) = 10 \frac{\mu\text{Sv}}{\text{h}}$ and still $t=150\text{min}$, the T , cooling time expected to reach this dose rate is equal to 93h.

⁹and also, for the copper BD, model overestimations

Bibliography

- [1] H.Wade Patterson, Ralph H. Thomas., *Accelerator Health Physics*, ACADEMIC PRESS, New York, London, (1973).
- [2] Gary J. Russell. *Spallation Physics - An Overview*, ICANS-XI International Collaboration on Advanced neutron Sources, KEK, Tsukuba, (October 22-26,1990).
- [3] A.H.Sullivan. *A Guide to Radiation and radioactivity Levels Near High Energy Particle Accelerators*, Nuclear technology Publishing, Ashford,Kent, TN23 1JW England, (1992).
- [4] R. Goloskie, K. Strauch, *Measurement of proton inelastic cross sections between 77 MeV and 133 MeV*, *Nuclear Physics*, **29**, p. 474-485, (January-February 1962).
- [5] R. G. P. Voss, R. Wilson, *Neutron Inelastic Cross-Sections between 55 and 140 MeV*, *The Royal Society*, volume 236, issue 1204,(10 July 1956)
- [6] G. R. Stevenson, *Induced activity in accelerator structures air and water*, Radiation Protection Dosimetry Vol. 96, No. 1-4,pp.373-380 (2011), Nuclear Technology Publishing.
- [7] K. Kitao, T. Ohata, *Gamma ray analysis on Radioactive Aerosol and Dust in the Machine Hall*, Health Physics, 21(1971).
- [8] S. Tavernier, *Experimental Techniques in Nuclear and Particle Physics*, Springer, Heidelberg, Dordrecht, London, New York, 2010.
- [9] Francesco Paolo La Torre , *Study of induced radioactivity in accelerator facilities*, PhD thesis, CERN .
- [10] Franck Goldembaum, *The physics of Spallation Processes*, ICTP, Trieste, 18 Oct. 2005.
- [11] A. Leuschner and K. Tesch, *The residual radioactivity of water copper beam dump for TESLA Test Facility*, Internal Report DESY d3-92, November 1998.
- [12] M. Lindroos, *Rewiew of ISOL-TYPE radioactive beam facilities*, Proceedings of EPAC 2004, Lucerne, Switzerland.

- [13] J.I.M. Botman and H.L. Hagedoorn ***EXTRACTION FROM CYCLOTRONS***,
Eindhoven University of Technology, Cyclotron Laboratory, Eindhoven, The Netherlands.